

The "normal" state of superconducting cuprates might really be normal after all

V. N. Zavaritsky^{1,2} and A. S. Alexandrov¹

¹Loughborough University, Loughborough, United Kingdom

²Kapitza Institute for Physical Problems & General Physics Institute, Moscow, Russia

High magnetic field studies of cuprate superconductors revealed a non-BCS temperature dependence of the upper critical field $H_{c2}(T)$ determined resistively by several groups. These determinations caused some doubts on the grounds of both the contrasting effect of the magnetic field on the in-plane and out-of-plane resistances reported for large Bi2212 sample and the large Nernst signal *well above* T_c . Here we present both $\rho_{ab}(B)$ and $\rho_c(B)$ of tiny Bi2212 crystals in magnetic fields up to 50 Tesla. None of our measurements revealed a situation when on the field increase ρ_c reaches its maximum while ρ_{ab} remains very small if not zero. The resistive $H_{c2}(T)$ estimated from $\rho_{ab}(B)$ and $\rho_c(B)$ are approximately the same. Our results support any theory of cuprates that describes the state above the resistive phase transition as perfectly normal with a zero off-diagonal order parameter. In particular, the anomalous Nernst effect above the resistive phase transition in high- T_c cuprates can be described quantitatively as a normal state phenomenon in a model with itinerant and localised fermions and/or charged bosons.

PACS numbers: 74.40.+k, 72.15.Jf, 74.72.-h, 74.25.Fy

A pseudogap is believed to be responsible for the non Fermi-liquid normal state of cuprate superconductors. Various microscopic models of the pseudogap proposed are mostly based on strong electron correlations [1], and/or on strong electron-phonon interaction[2]. There is also a phenomenological scenario [3], where the superconducting order parameter (the Bogoliubov-Gor'kov anomalous average $F(\mathbf{r}, \mathbf{r}') = \langle \psi_\downarrow(\mathbf{r})\psi_\uparrow(\mathbf{r}') \rangle$) does not disappear at T_c but at much higher (pseudogap) temperature. While the scenario [3] was found to be inconsistent with the 'intrinsic tunnelling' I-V characteristics, the discovery of the Joule heating origin of the gap-like I-V nonlinearities made that objection irrelevant [4]. Some other measurements [5] also provide evidence in support of [3].

In line with the scenario, several authors [6, 7] suggested a radical revision of the magnetic phase diagram of the cuprates with an upper critical field much higher than the resistive $H_{c2}(T)$. In particular, Ref.[6] questioned the resistive determination of $H_{c2}(T)$ [8, 9], claiming that, while ρ_c measures the inter-plane tunnelling, only the in-plane data represent a true normal state. The main argument in favour of this claim came from the radically different field dependencies of ρ_c and ρ_{ab} in Ref.[6] (shown below in our Fig.2B). According to this finding, a magnetic field sufficient to recover the normal state ρ_c , leaves in-plane superconductivity virtually unaffected. This discrepancy suggests that Bi2212 crystals do not lose their off-diagonal order in CuO₂ planes even well above $H_{c2}(T)$ determined from $\rho_c(B, T)$. However, this conclusion is based on one measurement and so certainly deserves experimental verification, which was not possible until recently because of the lack of reliable $\rho_{ab}(B, T)$ for Bi2212.

Quite similar conclusions followed from thermomagnetic studies of superconducting cuprates. Here a large Nernst signal *well above* T_c has been attributed to a *vortex* motion. As a result, the magnetic phase diagram of

the cuprates has been revised radically. Most surprisingly, Ref.[7] estimated H_{c2} at the zero-field transition temperature, T_{c0} , of Bi2212 as high as 50-150 Tesla.

On the other hand, any scenario with $F(\mathbf{r}, \mathbf{r}') \neq 0$ in the "normal" state is difficult to reconcile with the extremely sharp resistive and magnetic transitions at T_c in single crystals of cuprates. Above T_c , the uniform magnetic susceptibility is paramagnetic and the resistivity is perfectly 'normal', showing only a few percent positive or negative magnetoresistance (MR). Both in-plane [10, 11, 12] and out-of-plane [8] resistive transitions remain sharp in the magnetic field in high-quality samples, providing a reliable determination of a genuine $H_{c2}(T)$. These and some other observations [13] do not support any *stationary* superconducting order parameter above T_c .

Resolution of these issues, which affect fundamental conclusions about the nature of superconductivity in highly anisotropic layered cuprates, requires further careful experiments and transparent interpretations. Here we present systematic measurements of both in-plane and out-of-plane MRs of small Bi2212 single crystals subjected to magnetic fields, $B \leq 50$ Tesla, $B \perp (ab)$. Our measurements reproduced neither the unusual field dependence of ρ_{ab} nor the contrasting effect of the field as in [6], which are most probably an experimental artefact. On the contrary, they show that $H_{c2}(T)$ estimated from ρ_{ab} and ρ_c are nearly identical. These results, along with a simple explanation of the unusual Nernst signal in cuprates as a normal state phenomenon [14], strongly support any microscopic theory of cuprates with a zero off-diagonal order parameter above resistive $T_c(B)$.

Reliable measurements of the resistivity tensor require defect-free samples. This is of prime importance for in-plane MR because, owing to the extreme anisotropy of Bi2212 [15], even unit-cell scale defects will result in a significant out-of-plane contribution. Not only are such

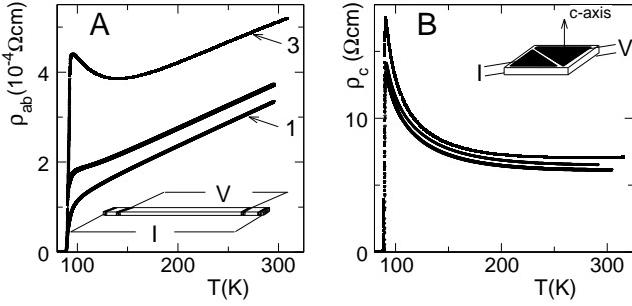


FIG. 1: Contact layout and examples of $\rho_c(T)$ and $\rho_{ab}(T)$ measured on small samples cleaved from the same Bi2212 crystal. ρ_c contamination of ρ_{ab} is $\sim 10^{-5}$ and indistinguishable for the curves labelled as 3 and 1 respectively.

minor defects impossible to detect by conventional techniques, but ρ_{ab} contamination with ρ_c might occur even in a perfect crystal with nonuniform current distribution. For these reasons, we paid special attention to sample preparation and selection [15]. Since the extremely high and temperature dependent electric anisotropy of Bi2212 prevents reliable measurement of both the in-plane and out-of-plane resistances on the same sample, we measured ρ_c and ρ_{ab} on different pieces of the same high-quality, optimally and slightly underdoped Bi2212 parent crystals with $T_{c0} \approx 87\text{--}92K$. As the specific demands of pulsed field experiments make it essential to use tiny specimens, we measured ρ_c on samples with in-plane dimensions from $\simeq 30 \times 30\mu\text{m}^2$ to $\simeq 80 \times 80\mu\text{m}^2$, while ρ_{ab} was studied on longer crystals, from $\simeq 300 \times 11\mu\text{m}^2$ to $\simeq 780 \times 22\mu\text{m}^2$. The samples for this study were selected on the basis of comparative analysis of transport measurements of 7-12 pairs of such samples, cleaved from different places of the same parent crystal (typically of $1 - 3\mu\text{m}$ thickness). To achieve a uniform *in-plane* current distribution, the current contacts were made by immersion of the crystals' ends into diluted conductive composite; ρ_c was measured with the contacts deposited on both ab-faces, see Fig.1. The uncertainty of the samples' dimensions is most probable cause of the mismatch of ρ_c in different pieces, Fig.1B. Unlike $\rho_c(T)$ curves, $\rho_{ab}(T)$ of different pieces often reveal qualitatively different behaviour, illustrated in Fig.1A. While the majority of the ' ρ_{ab} -samples' had the metallic type of zero-field $\rho_{ab}(T)$ represented by the curve 1, others demonstrated the sample-dependent $\rho_{ab}(T)$ upturn, which we attribute to ρ_c contamination. Only the samples with the lowest $\rho_{ab}(T)$ were selected for this study. The metallic type of zero-field $\rho_{ab}(T)$ and the *sign* of its normal state MR [15] indicate a vanishing ρ_c -contribution. The absence of hysteresis in the $\rho(B)$ data obtained on the rising and falling sides of the pulse and the consistency of $\rho(B)$ taken at the same temperature in pulses of different B_{max} exclude any measurable heating effects. The Ohmic response is confirmed by the consistency of $\rho(B)$ measured at identical conditions with different currents, $10\text{--}1000A/cm^2$ for

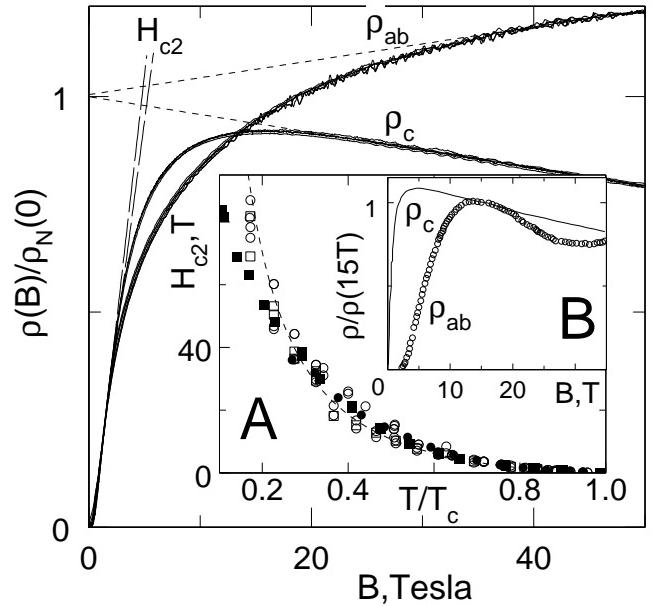


FIG. 2: $\rho_c(B)$ and $\rho_{ab}(B)$ of Bi2212 at $\sim 68K$, normalised by corresponding $\rho_N(0, T)$ obtained with the linear extrapolation from the normal state region (short dashes). The linear fits, shown by long dashed lines, refer to the flux-flow region. Inset A: H_{c2} estimated from $\rho_{ab}(B)$ and $\rho_c(B)$ is shown by the open and solid symbols respectively together with the fit, $H_{c2}(T) \sim (t^{-1} - t^{1/2})^{3/2}$, with $t = T/T_c$ [21] (broken line). Inset B shows ρ_c and ρ_{ab} from Ref.[6].

ρ_{ab} and $0.1\text{--}20A/cm^2$ for ρ_c .

Fig.2 shows the typical $\rho_c(B)$ and $\rho_{ab}(B)$ taken below T_{c0} of a Bi2212 single crystal. The low-field portions of the curves correspond to the resistance driven by vortex dynamics. Here, a non-linear $\rho(B)$ dependence is followed by a regime in which linear dependence fits the experimental observations rather well, Fig.2. It is natural to attribute the high field portions of the curves in Fig.2 (assumed to be above H_{c2}) to a normal state [9]. Here, the c-axis high-field MR appears to be negative and quasi-linear in B in a wide temperature range both above and below T_{c0} . Contrary to $\rho_c(B)$, the normal state in-plane MR is *positive* (see [15] and references therein for an explanation). The resistive upper critical field, $H_{c2}(T)$, is estimated from $\rho_c(B)$ and $\rho_{ab}(B)$ either as the intersection of two linear approximations in Fig.2, or from the flux-flow resistance as $H_{c2} = \rho_N(0, T)(\partial\rho_{FF}/\partial B)^{-1}$; both estimates are found to be almost identical. This procedure allows us to separate contributions originating from the normal and superconducting states and, in particular, to avoid ambiguity resulting from fluctuations in the crossover region. The downward deviations from the linear field dependence at fields around H_{c2} in Fig.2 are most likely caused by the conventional (3D-XY [16]) critical behaviour rather than the stationary off-diagonal order parameter in the "normal" phase [17]. The reasonable concordance of $H_{c2}(T)$ estimates from $\rho_c(B)$ and

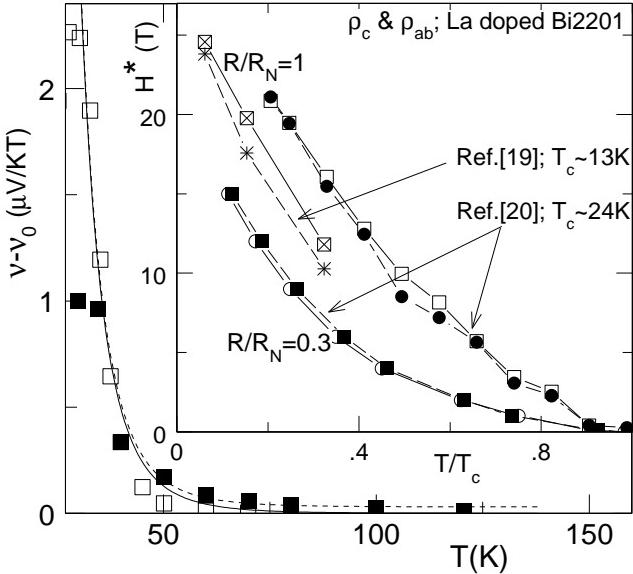


FIG. 3: \square and \blacksquare represent $\nu = e_y/B$ measured on Bi2201 [28] and YBCO [29] respectively; the solid and broken lines show fit with $a \propto T^{-6} - \nu_0$ for $\nu_0=0$ and $-0.03\mu V/KT$ respectively. Inset: $H_{c2}(T)$ obtained from the independent resistive studies of Bi2201 [19, 20]; the broken and solid lines correspond to the data taken from ρ_c and ρ_{ab} respectively.

$\rho_{ab}(B)$ (Fig.2A) favours our association of the resistive H_{c2} with the upper critical field, especially given the apparently different mechanisms responsible for ρ_{ab} and ρ_c [15].

Our conclusion is based on the results obtained during several hundred measurements performed on three pairs of crystals. None of those revealed a situation in which on field increase ρ_c reaches its maximum while ρ_{ab} remains very small if not zero as in [6] (see Fig.2B). Since the authors of Ref.[6] measured ' $\rho_{ab}(B)$ ' by means of contacts situated on the same face of the crystal while the current was injected into the opposite face, their curve could *not* represent the true ρ_{ab} . We cannot exclude the possibility that this observation might be caused by current redistribution in the medium with field and temperature dependent anisotropy. This opinion is supported by the independent study of current redistribution in homogeneous Bi2212, [18]. However, the threefold ρ_c enhancement warrants inhomogeneity of the huge crystal in [6] so that the results of [18] may not be directly applicable to this case. Neither the current redistribution nor imperfections of the crystal were accounted for in [6].

Our conclusions are supported by independent studies of a single-layer cuprate Bi(La)2201 with similar anisotropy. If we apply the routine procedure for the $H_{c2}(T)$ evaluation[8], very similar values of $H_{c2}(T)$ are obtained from ρ_{ab} and ρ_c measured on *the same* crystals [19] and films [20] (see broken and solid lines in the inset to Fig.3). The functional similarity of $H_{c2}(T)$ dependences estimated for the same conditions from resistivi-

ties of physically different origin is evident from Fig.2A and Fig.3(inset). Remarkably, these $H_{c2}(T)$ are compatible with the Bose-Einstein condensation field of pre-formed charged bosons [21] (Fig.2A), and also with some other models [22]. The described experiments were performed in optimally doped or only slightly underdoped samples. It would be desirable to extend these studies to more underdoped samples, where the conditions for bosonic superconductivity [2] are definitely satisfied.

Finally, we briefly address the origin of the Nernst effect in *superconducting* cuprates, which is found to be enormous well above T_c , in drastic contrast with conventional superconductors. While a significant fraction of research in the field of high-temperature superconductivity [23] describes the unusual Nernst signal as a signature of a nonzero superconducting order parameter above (resistive) T_c , a key to resolution of this dichotomy lies most likely in a qualitatively different normal state of cuprates as compared with conventional superconductors. While the latter are reasonably good metals, cuprates are known to be non-stoichiometric compounds. Moreover, undoped cuprates are insulators and their (super)conductivity appears as a result of doping, which inevitably introduces additional disorder. For these reasons, the conventional theory of heavily doped semiconductors and disordered metals might provide an adequate description of the normal state kinetic properties of cuprates (see [14] for more details). Carriers in doped semiconductors occupy states localised by disorder and itinerant Bloch-like states. Both types of carriers contribute to transport properties if the chemical potential μ (or the Fermi level) is close to the energy at which the lowest itinerant state appears (i.e. the mobility edge). When the chemical potential is near the mobility edge, and the effective mass approximation is applied, there is no Nernst signal from itinerant carriers alone because of the so-called Sondheimer cancellation [24]. However, when localised carriers contribute to the longitudinal transport, a finite positive Nernst signal $e_y \equiv -E_y/\nabla_x T$ appears as [14]

$$\frac{e_y}{\rho} = \frac{k_B}{e} r \theta \sigma_l, \quad (1)$$

where $\rho = 1/[(2s+1)\sigma_{xx}]$ is the resistivity, s is the carrier spin, r is nearly constant ($r \approx 14.3$ for fermions $s=1/2$, $r \approx 2.4$ for bosons $s=0$), and Θ is the Hall angle. Here, σ_{xx} is the conductivity of itinerant carriers, and σ_l is the conductivity of localised carriers, which obeys Mott's law, $\sigma_l = \sigma_0 \exp[-(T_0/T)^{1/3}]$. In two dimensions $T_0 \approx 8\alpha^2/(k_B N_l)$, where N_l is the DOS at the Fermi level[25, 26, 27].

In a sufficiently strong magnetic field, the radius of the 'impurity' wave function α^{-1} is about the magnetic length, $\alpha \approx (eB)^{1/2}$. Then the Nernst signal is given by

$$\frac{e_y}{B\rho} = a(T) \exp[-b(B/T)^{1/3}], \quad (2)$$

where $a(T) \propto T^{-6}$ [14] and $b = 2[e/(k_B N_l)]^{1/3}$ is a con-

stant determined by the density of impurities. As follows from Eq.(2), $a(T)$ is mostly responsible for the temperature dependence of the Nernst voltage above $T_c(B)$. Interestingly, the temperature dependences of the experimental Nernst voltage taken at fixed field, e_y/B , in LSCO, YBCO, Bi2212, and Bi(La)2201 single crystals agree reasonably with this theoretical result, as is illustrated in Fig.3 for YBCO-0.45 and Bi(La)2201-0.4. Although this qualitative agreement favours our model, convincing verification of the theory would be provided by analysis of the complementary $e_y(B, T)$ and $\rho(B, T)$ 2D-arrays of experimental data. This decisive comparison was recently performed. Remarkably, we found that the single-parameter relation, Eq.(2), *quantitatively* describes both the *field and temperature* dependencies of $e_y/(B\rho)$ measured experimentally above the resistive critical temperature $T_c(B)$ (see Ref.[14] for more details). Thus we conclude that the simple model with itinerant and localised fermions and/or charged bosons is compatible with most significant thermomagnetic and kinetic measurements in superconducting cuprates.

To conclude, we have shown that reliable experimental data do not require radical revision of the magnetic phase diagram of cuprates [30]. In particular, the reasonable concordance of resistive upper critical fields estimated from $\rho_{ab}(B)$ and $\rho_c(B)$ favours our assignment of resistive H_{c2} to the genuine upper critical field, especially given the apparently different mechanisms responsible for the in-plane and out-of-plane resistivity in the normal state of Bi2212 and Bi(La)2201, as evidenced by the huge and temperature dependent anisotropy, $\rho_c/\rho_{ab} \geq 10^4 - 10^5$. Our experimental $\rho_{ab}(T, B)$ and $\rho_c(T, B)$ in the same Bi2212 crystals and the model of the Nernst signal support virtually any microscopic theory that describes the state above the resistive and magnetic phase transition in superconducting cuprates as perfectly 'normal' with $F(\mathbf{r}, \mathbf{r}') = 0$. The carriers could be normal-state fermions, as in any BCS-like theory of cuprates, normal-state charged bosons, as in the bipolaron theory [2], or a mixture of both.

This work was supported by the Leverhulme Trust (grant F/00261/H).

-
- [1] for a review see T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999)
- [2] A.S. Alexandrov, *Theory of superconductivity* (IoP Publishing, Bristol-Philadelphia, 2003)
- [3] V.J. Emery, S.A. Kivelson, Nature, **374**, 434 (1995)
- [4] V.N. Zavaritsky, Physica C**404**, 440 (2004)
- [5] J. Corson, R. Mallozzi, J. Orenstein, J. N. Eckstein, I. Bozovic, Nature **398**, 221 (1999); C. Meingast, V. Pasler, P. Nagel, A. Rykov, S. Tajima, P. Olsson, Phys. Rev. Lett. **86**, 1606 (2001)
- [6] N.Morozov, L.Krusin-Elbaum, T.Shibauchi, L.N.Bulaevskii, M.P.Maley, Yu.I.Latyshev, T.Yamashita,Physica C **341-348**, 1511 (2000); Phys.Rev. Lett. **84**,1784 (2000)
- [7] Y. Wang, S. Ono, Y. Onose, G. Gu, Y. Ando, Y. Tokura, S. Uchida, N.P. Ong, Science **299**, 86 (2003)
- [8] A.S. Alexandrov, V.N. Zavaritsky, W.Y. Liang, P.L. Nevesky, Phys. Rev. Lett. **76** 983 (1996)
- [9] V.N. Zavaritsky, M. Springford, JETP Lett. **68**, 448 (1998); V.N.Zavaritsky, JETP Lett. **71**, 80 (2000)
- [10] A.P. Mackenzie, S.R. Julian, G.G. Lonzarich, A. Carrington, S.D. Hughes, R.S. Liu, D.C. Sinclair, Phys. Rev. Lett. **71**, 1238 (1993)
- [11] M.S. Osofsky, R. J. Soulen, S.A. Wolf, J.M. Broto, H. Rakoto, J.C. Ousset, G. Coffe, S. Askenazy, P. Pari, I. Bozovic, J.N. Eckstein, G. F. Virshup, Phys. Rev. Lett. **71**, 2315 (1993); ibid **72**, 3292 (1994)
- [12] D.D. Lawrie, J.P. Franck, J.R. Beamish, E.B. Molz, W.M. Chen, M.J. Graft, J. Low Temp. Phys. **107**, 491 (1997)
- [13] J. W. Loram, K. A. Mirza, J.R. Cooper, W.Y. Liang, J.M. Wade, J. Superconductivity, **7**, 243 (1994)
- [14] A.S.Alexandrov, V.N.Zavaritsky, cond-mat/0401272
- [15] V.N. Zavaritsky, J.Vanacen, V.V.Moshchalkov, A.S. Alexandrov, Physica C(Amsterdam) **404**, 444 (2004); cond-mat/0308256
- [16] T. Schneider, J.M. Singer, *Phase Transition Approach to High Temperature Superconductivity* (Imperial College Press, London, 2000)
- [17] As is clear from Fig.2 the deviation in $\rho_{ab}(B)$ occurs at fields somewhat higher than in $\rho_c(B)$. This could be explained within a phenomenological model of preformed pairs (bosons) by critical enhancement of the relaxation time of bosons near their Bose-Einstein condensation temperature, $T_c(B)$, $\tau \propto [T - T_c(B)]^{-1/2}$, as in atomic Bose-gases, eg. T. Lopez-Arias, A. Smerzi, Phys.Rev. A **58**, 526 (1998)
- [18] R.Busch, G.Ries, H.Werthner, G.Kreiselmeyer, G.Saemann-Ischenko, Phys. Rev. Lett. **69**, 522 (1992)
- [19] Y.Ando, G.S.Boebinger, A.Passner, N.L.Wang, C.Geibel, F.Steglich, Phys.Rev. Lett. **77**, 2065 (1996)
- [20] Y.Z.Zhang, R.Deltour, J.-F. de Marneffe, Y.L.Qin, L.Li, Z.X.Zhao, A.G.M.Jansen, P.Wyder, Phys. Rev. B**61**, 8675 (2000)
- [21] A.S. Alexandrov, Phys. Rev. B **48**, 10571 (1993)
- [22] A.A. Abrikosov, Phys. Rev. B **56**, 446 (1997); D.S. Fisher, M.P.A. Fisher, D.A. Huse, *ibid* **43**,130 (1991); Y.N.Ovchinnikov, V.Z.Kresin, *ibid* **54**,1251 (1996)
- [23] I. Ussishkin, S.L. Sondhi, D.A. Huse, Phys. Rev. Lett. **89**, 287001 (2002); C. Honerkamp, P.A. Lee, *ibid* **92**, 177002 (2004); S. Mukerjee, D.A. Huse, cond-mat/0307005; S.Tan, K.Levin, Phys.Rev. B **69**, 064510 (2004)
- [24] E.H. Sondheimer, Proc. Roy. Soc. **193**, 484 (1948)
- [25] N.F. Mott and E.A. Davis, *Electronic Processes in Non-Crystalline Materials* (Clarendon Press,Oxford, 1979).
- [26] B.I. Shklovskii, Soviet Phys. JETP Lett. **36**, 51 (1982)
- [27] R. Mansfield, H. Tokumoto, Phil Mag. B**48**, L1 (1983)
- [28] Y. Wang, Z.A. Xu, T. Kakeshita, S. Uchida, S. Ono, Y. Ando, N. P. Ong, cond-mat/0108242.
- [29] N. P. Ong, Y. Wang, S. Ono, Y. Ando, S. Uchida, cond-mat/0312213.
- [30] V.N. Zavaritsky, V.V. Kabanov, A.S. Alexandrov, Europhys. Lett. **60**, 127, (2002) and references therein.